1. DEFINITIONS AND COMPUTATIONS

This document describes the Fourier component matrices $\boldsymbol{R}^{m}(\mu,\mu')$ for the reflection matrix $\boldsymbol{R}(\mu,\mu',\varphi-\varphi')$ of ocean systems. Reflection matrix $\boldsymbol{R}(\mu,\mu',\varphi-\varphi')$ is defined as in Hansen and Travis (1974). That is, let skylight illuminate the ocean in directions defined by the polar zenith angle θ' and polar azimuth angle φ'. Also, let $\mu' = \cos\theta'$. Then

$$\boldsymbol{I}(\mu,\varphi) = \frac{1}{\pi}\int_0^1 \mu' d\mu' \int_0^{2\pi} d\varphi' \boldsymbol{R}(\mu,\mu',\varphi-\varphi')\boldsymbol{I}'(\mu',\varphi') \tag{1}$$

where $\boldsymbol{I}'$ and $\boldsymbol{I}$ contain Stokes parameters I, Q, and U of the skylight and of the light reflected by the ocean, respectively. Hence $\boldsymbol{R}(\mu,\mu',\varphi-\varphi')$ and $\boldsymbol{R}^{m}(\mu,\mu')$ are (3×3) matrices that ignore the circular polarized component of light described by Stokes parameter V. Stokes parameters Q and U are defined here with respect to the local meridional plane, i.e., the local plane containing the vertical axis and the direction of light propagation.

The Fourier component matrices $\boldsymbol{R}^{m}(\mu,\mu')$ follow the Fourier expansion given by Eq. (47) in De Haan *et al.* (1987), i.e.

$$\boldsymbol{R}(\mu,\mu',\varphi-\varphi') = \frac{1}{2}\sum_{m=0}^{M}(2-\delta_{m,0})\left\{\boldsymbol{B}^{+m}(\varphi-\varphi')\boldsymbol{R}^{m}(\mu,\mu')(\boldsymbol{1}+\boldsymbol{\Delta}) + \boldsymbol{B}^{-m}(\varphi-\varphi')\boldsymbol{R}^{m}(\mu,\mu')(\boldsymbol{1}-\boldsymbol{\Delta})\right\} \tag{2}$$

where $\boldsymbol{1}$ is the (3×3) unit matrix, and

$$\boldsymbol{B}^{+m}(\varphi) = diag(\cos m\varphi, \cos m\varphi, \sin m\varphi), \tag{3}$$

$$\boldsymbol{B}^{-m}(\varphi) = diag(-\sin m\varphi, -\sin m\varphi, \cos m\varphi)\ , \tag{4}$$

$$\boldsymbol{\Delta} = diag(1,1,-1)\ . \tag{5}$$

Furthermore, the $\boldsymbol{R}^{m}(\mu_i,\mu_j)$ matrices for all μ_i,μ_j values (i, j = 1, 2, 3, … , n) are stored together as 1 large ($3n$×$3n$) matrix $\boldsymbol{R}^{m}$ with

$$\boldsymbol{R}^{m}_{3(i-1)+l,3(j-1)+k} = \boldsymbol{R}^{m}_{lk}(\mu_i,\mu_j)\ . \tag{6}$$

Hence $\boldsymbol{R}^{m}$ is similar to the so-called supermatrix $\boldsymbol{L}^{m}$ defined by Eq. (83) in De Haan *et al.* (1987) except for omitting the weights $(2w_i,\mu_i)^{1/2}$ and $(2w_j,\mu_j)^{1/2}$, and for using the (3×3) approximation for scattering of polarized light.

The $\boldsymbol{R}(\mu,\mu',\varphi-\varphi')$ matrix is computed for an ocean system consisting of *(i)* a wind-ruffled ocean surface; and *(ii)* a scattering ocean body. Note that in this work $\boldsymbol{R}(\mu,\mu',\varphi-\varphi')$ describes only the *diffuse* radiance reflected by this ocean system, i.e. the skylight radiance transmitted by the ocean surface and scattered by the underlying ocean body. Hence, it does *not* include the specular sunglint radiance (i.e. the specular reflection of skylight radiance by the ocean surface). The reflection matrix for the specular sunglint radiance must still be added to the $\boldsymbol{R}(\mu,\mu',\varphi-\varphi')$ matrix provided by this work to obtain the complete reflection matrix for the ocean system.

The transmission of light by the wind-ruffled ocean surface is computed by applying the geometrical optics approach to the isotropic surface slope distribution from Cox and Munk (1954). The assumed refractive index is 1.34 and the assumed windspeed is 7 m/s for these computations. The surface does not contain foam, but surface shadowing effects are taken into account using the shadowing function from Sancer (1969). The resulting surface transmission matrices are normalized such that no energy is lost due to shadowing while preserving the symmetry relations given by Hovenier (1969). A more complete description for the ocean surface transmission matrix computations is given by Chowdhary (1999).

The ocean body for the underwater light scattering is assumed to be homogeneous and to have no bottom below. The ocean body has an optical thickness of 20 for all computations, and is assumed to be a homogeneous blend of pure sea water mixed with marine particulate and dissolved matter. Inelastic scattering sources such as fluorescence and Raman scattering are ignored. The corresponding (3×3) bulk ocean scattering matrix $\boldsymbol{F}_{\mathrm{blk}}(\lambda,\Theta)$ and single scattering albedo $\omega_{\mathrm{blk}}(\lambda)$, where λ is the wavelength Θ is the scattering angle, are computed from

$$\boldsymbol{F}_{\mathrm{blk}}(\Theta,\lambda)=\frac{b_{\mathrm{w(}}(\lambda)\boldsymbol{F}_{\mathrm{w}}(\Theta)+b_{\mathrm{p(}}(\lambda)\boldsymbol{F}_{\mathrm{p}}(\Theta)}{b_{\mathrm{w(}}(\lambda)+b_{\mathrm{p(}}(\lambda)} \tag{7}$$

and

$$\omega_{\mathrm{blk}}(\lambda)=\frac{b_{\mathrm{w}}(\lambda)+b_{\mathrm{p}}(\lambda)}{b_{\mathrm{w}}(\lambda)+b_{\mathrm{p}}(\lambda)+a_{\mathrm{blk}}(\lambda)}\ , \tag{8}$$

where $\boldsymbol{F}_{\mathrm{w}}(\Theta)$ and $\boldsymbol{F}_{\mathrm{p}}(\Theta)$ are the (3×3) scattering matrices for pure sea water and particulate matter, and where $b_{\mathrm{w}}(\lambda)$ and $b_{\mathrm{p}}(\lambda)$ are the corresponding scattering coefficients, respectively. Furthermore, $a_{\mathrm{blk}}(\lambda)$ is the absorption coefficient for bulk oceanic water, i.e. it is the sum of absorption coefficients for pure sea water, marine particulate matter, and marine dissolved matter. Matrices $\boldsymbol{F}_{\mathrm{w}}(\Theta)$ and $\boldsymbol{F}_{\mathrm{p}}(\Theta)$ are obtained following the work by Morel (1974) and by Chowdhary *et al.* (2006, 2012), respectively. Specifically, Rayleigh scattering law with a depolarization factor of 0.039 is assumed to compute $\boldsymbol{F}_{\mathrm{w}}(\Theta)$, whereas plankton and detritus particulates are mixed as a function of [Chl] to compute $\boldsymbol{F}_{\mathrm{p}}(\Theta)$. The values for $b_{\mathrm{w}}(\lambda)$ are tabulated in Smith and baker (1981), while $b_{\mathrm{p}}(\lambda)$ and $a_{\mathrm{blk}}(\lambda)$ are computed following Morel and Maritorena (2001) and Morel *et al.* (2007), respectively. Note that Huot *et al.* (2008) provide a correction for the spectrum of $b_{\mathrm{p}}(\lambda)$ while the website provided Morel *et al.* (2007) for the diffuse downward irradiance data to compute $a_{\mathrm{blk}}(\lambda)$ is defunct. Hence, the results for $b_{\mathrm{p}}(\lambda)$ and $a_{\mathrm{blk}}(\lambda)$ are provided in Table 1 as a function of λ and [Chl].

Table 1. Underwater light scattering and absorption coefficients §

coefficient	[Chl] ¶	λ = 380	λ = 385	λ = 410	λ = 440	λ = 470	λ = 550	λ = 670
$b_p(\lambda)$ §	0.03	0.0391	0.0387	0.0365	0.0342	0.0322	0.0279	0.0233
	0.10	0.0852	0.0844	0.0810	0.0774	0.0742	0.0670	0.0588
	0.30	0.1732	0.1723	0.1678	0.1630	0.1587	0.1487	0.1371
	1.00	0.3770	0.3761	0.3727	0.3688	0.3651	0.3566	0.3462
	3.00	0.8050	0.8050	0.8050	0.8050	0.8050	0.8050	0.8050
$a_{blk}(\lambda)$ §	0.03	0.0180	0.0167	0.0141	0.0144	0.0169	0.0571	0.4099 ‡
	0.10	0.0316	0.0298	0.0268	0.0259	0.0261	0.0634	0.4181 ‡
	0.30	0.0581	0.0554	0.0505	0.0477	0.0439	0.0730	0.4307 ‡
	1.00	0.1209	0.1160	0.1045	0.0984	0.0842	0.0911	0.4550
	3.00	0.2435	0.2348	0.2059	0.01953	0.1609	0.1211	0.4958

§ Units: m^{-1}; ¶ Units: mg/m^3; ‡ $a_w(\lambda)$ values from Pope and Fry (1997)

Multiple scattering computations for the underwater light are performed using the doubling/adding method described by de Haan *et al*. (1987), except for *not* separating the first order scattering contribution to reduce the M+1 number of Fourier terms in Eq. (2) for a pre-defined accuracy ε. Instead, the criterion used for terminating the Fourier series expansion of $\boldsymbol{R}(\mu,\mu',\varphi-\varphi')$ is that the resulting accuracy for the reflectance of $\boldsymbol{I}(\mu,\varphi)$ in Eq. (1), when seen by an observer just above the ocean surface, is better than ε. The skylight illumination $\boldsymbol{I}(\mu',\varphi')$ chosen for this purpose is the direct unidirectional sunlight attenuated by a purely molecular scattering atmosphere. Let τ_{mol} denote the optical thickness for this atmosphere. The criterion can then be written as

$$\left| \boldsymbol{R}^{m}_{(p,1)}(\mu_i,\mu_j)\exp(-\tau_{mol}/\mu_j) \right| < \varepsilon, \quad m \geq M+1 \tag{9}$$

to be satisfied $p \in \{1,2,3\}$ and for all pairs of (μ_i,μ_j). In this work, ε is set to 2×10^{-6}, τ_{mol} is taken from Hansen and Travis (1974), and the criterion in Eq. (9) is checked for m=M and m=M+1.

The source code for the radiative transfer (RT) computations is written in standard FORTRAN 77 language. It was originally developed by Drs. Victor L. Dolman at the Free

University in Amsterdam to compute elastic scattering of polarized light in plan-parallel, vertically inhomogeneous atmospheres (with an optional lower Lambertian boundary). Chowdhary (1991) extended the source code to include a smooth water surface (with an optional lower Lambertian boundary). The smooth water surface was later replaced by an ocean system consisting of a wind-ruffled water-surface and a plan-parallel, vertically inhomogeneous ocean body (with an optional lower Lambertian boundary) (Chowdhary, 1999). To compute the $\boldsymbol{R}^{\mathrm{m}}$ matrices in this work, the RT source code was compiled using the GNU Fortran Version 4.7.2 compiler. The following flags were used for this compilation:

```
-O2 -fbounds -fno-automatic -frecord-marker=4
```

Note that the `-frecord-marker=4` flag must then also be used to compile the FORTRAN code used to retrieve the stored $\boldsymbol{R}^{\mathrm{m}}$ matrices.

2. NOMENCLATURE, DATA, AND FORMAT OF RMocean FILES

The RT source code writes each computed $\boldsymbol{R}^{\mathrm{m}}$ matrix to an output file, denoted hereafter as an `RMocean` file. The nomenclature for the `RMocean` files is as follows:

```
RMocean_new_[wavelength]_C[Chl]_M[F-index]
```

where

`[wavelength]`	∈ {`380`, `385`, `410`, `440`, `470`, `490`, `550`, `670`}	is wavelength λ (nm),
`[Chl]`	∈ {`003`, `010`, `030`, `100`, `300`}	is Chlorophyll *a* amount [Chl] (10^{-2} mg/m^3)
`[F-index]`	∈ {`00`, `01`, `02`, … , `14`, `15`}	is Fourier index m in Eqs. (2)–(6).

Hence, there are in total 1,024 `RMocean` files. The FORTAN syntax used in the RT source code to write data to each of these files is as follows:

```
implicit double precision (a-h,o-z)
parameter (ndmu=60, ndsup=240)
dimension xmu(ndmu), Rm(ndsup,ndsup)
                   …
write(ifile) nmug, nmutot, nmat, lambda, Chl, Ir3x
write(ifile) xmu
write(ifile) Rm
```

where `ifile` is an internal source code number that identifies each `RMocean` file, and where

`nmug`	=	the number of Gaussian quadrature points used for the RT computations,
`nmutot`	=	the total number n of μ' points used for μ_i and μ_j in Eq. (6),
`nmat`	=	the number of Stokes parameters considered in the RT computations,

`lambda` = the wavelength λ (nm).
`Chl` = the Chlorophyll *a* concentration [Chl] (mg/m^3),
`Irx` = identifies whether $\boldsymbol{R}^{\mathrm{m}}$ is non-zero (`Irx`=1) or set to zero (`Irx`=0),
`xmu` = the array containing the values and order of μ' used for μ_i and μ_j in Eq. (6),
`Rm` = the $\boldsymbol{R}^{\mathrm{m}}$ matrix.

In this work, `nmug`=30, `nmutot`=60, `nmat`=3, and `xmu` contains the Gaussian quadrature points μ_i (i = 1, …, `nmug`) plus the extra user-defined points μ_i (i = `nmug`+1, … , `nmutot`) that are given in Table 2. Finally, `Irx` =1 if $m \leq M$+1 in Eq. (9), and `Irx`=0 if otherwise.

Table 2. Gaussian quadrature and user-defined points for skylight directions θ_i and μ_i.

i	θ_i (deg)	$\mu_i = \cos\theta_i$	i	θ_i (deg)	$\mu_i = \cos\theta_i$	i	θ_i (deg)	$\mu_i = \cos\theta_i$	i	θ_i (deg)	$\mu_i = \cos\theta_i$
1	89.9110	0.001553	16	58.2822	0.525736	31	23.8000	0.999848	46	34.0000	0.829038
2	89.5321	0.008166	17	54.7647	0.576935	32	3.2000	0.998441	47	36.2000	0.806960
3	88.8546	0.019989	18	51.1474	0.627318	33	5.4000	0.995562	48	38.4000	0.783693
4	87.8853	0.036900	19	47.4407	0.676352	34	7.6000	0.991216	49	40.6000	0.759271
5	86.6337	0.058720	20	43.6544	0.723517	35	9.8000	0.985408	50	42.8000	0.733730
6	85.1115	0.085217	21	39.7974	0.768312	36	12.0000	0.978148	51	45.0000	0.707107
7	83.3323	0.116111	22	35.8784	0.810263	37	14.2000	0.969445	52	47.2000	0.679441
8	81.3108	0.151075	23	31.9050	0.848925	38	16.4000	0.959314	53	49.4000	0.650774
9	79.0626	0.189737	24	27.8849	0.883889	39	18.6000	0.947768	54	51.6000	0.621148
10	76.6035	0.231688	25	23.8251	0.914783	40	20.8000	0.934826	55	53.8000	0.590606
11	73.9496	0.276483	26	19.7323	0.941280	41	23.0000	0.920505	56	56.0000	0.559193
12	71.1163	0.323648	27	15.6133	0.963100	42	25.2000	0.904827	57	58.2000	0.526956
13	68.1189	0.372682	28	11.4752	0.980011	43	27.4000	0.887815	58	60.4000	0.493942
14	64.9718	0.423065	29	7.3272	0.991834	44	29.6000	0.869495	59	62.6000	0.460200
15	61.6886	0.474264	30	3.1939	0.998447	45	31.8000	0.849893	60	64.8000	0.425779

Furthermore, `Irx` =1 if $m \leq M+1$ in Eq. (9), and `Irx`=0 if otherwise.

REFERENCES

Chowdhary, J. (1991), "*Incorporation of a smooth water-air interface in multiple scattering calculations using the adding method for polarized light*," M.A. Thesis, Free University, Amsterdam.

Chowdhary, J. (1999), "*Multiple scattering of polarized light in atmosphere-ocean systems*," Ph. D. Thesis, Columbia University, New York.

Chowdhary, J., Cairns, B., and Travis, L. D. (2006), "Contribution of waterleaving radiances to multiangle, multispectral polarimetric observations over the open ocean: bio-optical model results for case 1 waters," *Applied Optics* **45**:5542−5567.

Chowdhary, J., Cairns, B., Waquet, F., Knobelspiesse, K., Ottaviani, M., Redemann, J., Travis, L., and Mishchenko, M. (2012), "Sensitivity of multiangle, multispectral polarimetric remote sensing over open oceans to water-leaving radiance: Analyses of RSP data acquired during the MILAGRO campaign." *Remote Sensing of Environment* **118**:284−308.

Cox, C., & Munk, W. (1954), "Statistics of the sea-surface derived from sun-glitter," *Journal of Marine Research* **13**:198−227.

de Haan, J. F., Bosma, P. B., and Hovenier, J. W. (1987), "The adding method for multiple scattering calculations of polarized light," *Astronomy and Astrophysics* **183**:371−391.

Hansen, J. E., and Travis, L. D. (1974), "Light scattering in planetary atmospheres," *Space Science Reviews* **16**:527–610.

Huot, Y., Morel, A., Twardowski, M. S., Stramski, D., and Reynolds, R. A. (2008), "Particle optical backscattering along a chlorophyll gradient in the upper layer of the eastern South Pacific Ocean,".*Biogeosciences* **5**:495−507.

Morel, A., and Maritorena, S. (2001), "Bio-optical properties of oceanic waters: a reappraisal," *Journal of Geophysical Research* **106**:7163−7180.

Morel, A., Claustre, H., Antoine, D., and Gentili, B. (2007), "Natural variability of bio-optical properties in Case 1 waters: attenuation and reflectance within the visible and near-UV spectral domains, as observed in South Pacific and Mediterranean waters," *Biogeosciences* **4**:913−925.

Pope, R. M., & Fry, E. S. (1997), "Absorption spectrum (380−700 nm) of pure water. II. Integration cavity measurements," *Applied Optics* **36**:8710−8723.

Sancer, M. I., (1969), "Shadow-corrected electromagnetic scattering from a randomly-rough ocean surface," *IEEE Transactions on Antennas and Propagation* **17**:557−585.

Smith, R. C., and Baker, K. S. (1981), "Optical properties of the clearest natural waters (200−800 nm)," *Applied Optics* **20**:177−184.